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## **JAPANESE / ENGLISH TRANSLATION OF**

**Japanese Patent Application JP 58 - 214208 A**

**Electric Insulating Film**

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**(54) Electric Insulating Film**

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**SPECIFICATION**

**1. Title of the Invention**

Electric insulating film

**2. Claims**

(1) Electric insulating film, comprising a polyalkylene terephthalate as a principal component and having the following characteristics (a) through (c).

- (a) Intrinsic viscosity of 0.50 dL/g or greater
- (b) Trichloroethylene equilibrium adsorption rate at 20°C of 15 wt% or less
- (c) Percentage dimensional change at 245°C of no greater than 5%

(2) The electric insulating film according to claim 1, characterized in that a metal layer is formed on at least one side of the electric insulating film.

### 3. Detailed Description of the Invention

The present invention relates to a novel insulating film, and, in particular, relates to an insulating film for flexible printed circuit base film or cover film, flat cables, and integrated circuit chip carrier tape.

In further detail, the present invention relates to an electric insulating film comprising a polyalkylene terephthalate as a principal component and having excellent properties; including mechanical properties, electrical properties, dimensional stability, heat resistance, and chemical resistance.

A conductive metal layer is conventionally formed on electric insulating film, such as polyethylene terephthalate biaxially oriented film or polyimide film, and the film with the metal layer is used for flexible printed circuits, flat cables, integrated circuit chip carrier tape, and the like. The demand for this type of film has increased with the development of electronic equipment that is smaller, lighter, and of higher density.

Although polyethylene terephthalate biaxially oriented film has excellent mechanical properties and electrical properties, it cannot be said that it has sufficient heat resistance. This film has a large coefficient of contraction even when heated during layering or soldering to a temperature that does not exceed the melting point, such as 200°C to 230°C, so use of the film is restricted considerably. Polyimide film has excellent mechanical properties and heat resistance, but its equilibrium moisture content is high, so it is necessary to closely control the moisture content during layering and soldering. Moreover, the reduction in electrical properties due to absorption of moisture is also a problem. In addition, the film must be produced by means of the solution method, bringing about higher costs.

The inventors focused on this point and completed the present invention after intense studies intended to produce an electric insulating film comprising a polyalkylene terephthalate as a principal component and possessing the melt moldability, excellent electrical properties, and moisture resistance of the polyalkylene terephthalate together with superb heat resistance and dimensional stability.

That is, the present invention is an electric insulating film that comprises a polyalkylene terephthalate as a principal component and has the following characteristics (a) through (c).

- (a) Intrinsic viscosity of 0.50 dL/g or greater
- (b) Trichloroethylene equilibrium adsorption rate at 20°C of 15 wt% or less
- (c) Percentage dimensional change at 245°C of no greater than 5%

The alkylene terephthalate component accounts for 60 wt% or more, preferably 80 wt% or more, and particularly preferably 90 wt% or more, of the electric insulating film that has a polyalkylene terephthalate as its principal component and is used in the present invention.

When the alkylene glycol residue constituting the alkylene terephthalate component is an alkylene glycol, it has two to ten carbons. Examples are ethylene glycol, trimethylene glycol, tetramethylene glycol, hexamethylene glycol, cyclohexanedimethanol, and neopentyl glycol. It is preferred that ethylene terephthalate or cyclohexanedimethylene terephthalate account for 70 wt% or more of this electric insulating film.

The alkylene terephthalate component having these glycols and terephthalic acid as its constituent components can be modified by means of copolymerization, blending, or the like within the aforementioned range.

The glycols or dicarboxylic acids illustrated below and their ester-forming derivatives may be used in the case of copolymerization. Examples of suitable glycols include polyalkylene glycols with 4 to 500 carbons, and N,N'-bis(hydroxyethyl)benzophenone tetracarboximide. Examples of dicarboxylic acids are aromatic dicarboxylic acids such as isophthalic acid, naphthalenedicarboxylic acid, diphenyldicarboxylic acid, diphenylsulfonedicarboxylic acid, diphenoxyethanedicarboxylic acid, and diphenyletherdicarboxylic acid; aliphatic dicarboxylic acids such as succinic acid, adipic acid, sebacic acid, and dodecanedioic acid; alicyclic acids such as cyclohexanedicarboxylic acid; and oxycarboxylic acids such as oxybenzoic acid and  $\epsilon$ -oxycaproic acid.

Polyester in which the aforementioned polyalkylene terephthalate is a principal component is obtained by means of the polycondensation of the aforementioned dicarboxylic

acid and/or the ester-forming derivative thereof, as well as the aforementioned glycol and/or the ester-forming derivative thereof. Conventional methods can be used for this production process. Moreover, polyfunctional compounds such as pentaerythritol, trimethylolpropane, pyromellitic acid, and trimellitic acid; or unsaturated compounds such as maleic acid, fumaric acid, tetrahydrophthalic anhydride, endo-bicyclo[2,2,1]-5-heptene-2,3-dicarboxylic anhydride, dimer acids, and ester-forming derivatives thereof can be copolymerized with the aforementioned polyester as long a molten film can be effectively formed.

Other resins can be used for modification as a result of blending. Examples are polyesters other than polyalkylene terephthalate, as well as polyarylene esters, polycarbonates, polysulfones, polyether sulfones, polyphenylene sulfides, polyester imides, polyamide imides, polyimides, silicone resins, and polyolefins; inorganic fillers such as silica, talc, kaolin, titanium oxide, and glass powder; and flame retarding agents, antioxidants, ultraviolet absorbers, crosslinking agents, and the like. Examples of suitable blending methods include methods of mechanical mixing using an S blender or V blender and then uniformly blending the components using a blender such as an extruder; methods of melting a polymer in a melt extruder, feeding metered amounts of the other components together with the measured stock of the molten polymer being fed, and blending specific amounts of the components in a molten state; and other conventional blending methods.

The electric insulating film of the present invention, in which a polyalkylene terephthalate serves as a principal component, may have a polyalkylene terephthalate homopolyester or copolyester as the principal component, as previously described. The intrinsic viscosity of the polyalkylene terephthalate or copolyester thereof as a principal component is 0.5 dL/g or greater, preferably 0.70 dL/g or greater, and ideally 0.80 dL/g or greater, as measured at 30°C in phenol/1,1,2,2-tetrachlorethane = 60/10 (weight ratio). There are no special restrictions as to the upper limit, but it is usually 2.0 dL/g or less, and ideally 1.5 dL/g or less. If the intrinsic viscosity is less than 0.50 dL/g, it will be difficult to manufacture a thin film by melt film formation, and it will be impossible to obtain the mechanical properties needed for an electric insulating film.

The trichloroethylene equilibrium adsorption rate at 20°C of the electric insulating film comprising a polyethylene terephthalate as a principal component in accordance with the present invention must be 1.5 wt% or less. If the rate exceeds 15 wt%, the film will not have the solvent resistance and heat resistance that are necessary to process and use the film for flexible printed circuits or the like, and the film will curl or crack, making it inappropriate for use as an electric insulating film. Polyalkylene terephthalate resin is inherently easily attacked by trichloroethylene, but it has been shown that resistance to trichloroethylene varies considerably depending on the microstructure of the film formed from this resin. The detailed correlation between the amount of trichloroethylene adsorption and the film microstructure is not known, but a polyalkylene terephthalate film with a trichloroethylene adsorption rate of 15 wt% or less, preferably 10 wt% or less, and particularly 5 wt% or less, has excellent properties as an electric insulating film in terms of solvent resistance and heat resistance because of its microstructure, of which the trichloroethylene adsorption rate is a characteristic.

Furthermore, the percentage dimensional change at 245°C of the electric insulating film of the present invention must be no greater than 5%. This percentage dimensional change reflects a structure whereby the film of the present invention has high dimensional stability in the presence of heat. A percentage that exceeds 5% is inappropriate because the film will crack or curl when exposed to heat during the production or use of a printed substrate.

The electric insulating film of the present invention is obtained by means of extruding a resin comprising as a principal component a polyalkylene terephthalate homopolyester or copolyester (including resin modified as a result of copolymerization or blending) in molten form from a flat die or a circular die, and cooling the extrudate on a roll in air, an inert gas, an inert liquid, or a combination thereof. The cooled extrudate is usually further heat-treated. The melt extrusion temperature is usually 250 to 360°C, preferably 260 to 330°C, and the heat treatment temperature is usually 130 to 260°C, preferably 150 to 240°C. The heat treatment is performed for the purpose of alleviating the warping or the extension or orientation of the molecular chain when the film is formed, and crystallizing the film. Moreover, precautions should be taken so that the warping or the extension and orientation of the molecular chain in the amorphous segments do not recur during heat treatment. The method whereby the film is crystallized in unaltered form while allowed to retain its natural dimensions (in a freely stretched

or contracted state) at the heat treatment temperature is used in order to accomplish this purpose. Specifically, a method whereby the product is tented in accordance with thermal expansion and is at the same time heated and crystallized using hot air, or the method of crystallization in a relaxed state on a roll is used to allow the film to retain its natural dimensions. The resulting film has essentially no extended chains in the amorphous segments, and is a substantially unoriented film. The degree of crystallization, as measured with the help of the X-ray method, is usually 5% to 70%.

The thickness of the electric insulating film of the present invention is usually 1  $\mu$  to 500  $\mu$ , preferably 3  $\mu$  to 350  $\mu$ , and particularly preferably 10  $\mu$  to 100  $\mu$ .

The electric insulating film of the present invention is often provided with a metal layer on at least one side. The metal layer can be formed on the aforementioned electric insulating film, directly or with an adhesive layer in between, using as needed (1) a method whereby metal foil is layered, (2) the method of electroless chemical plating, (3) evaporation, sputtering, or ion plating, (4) a method that is a combination of these with electroplating, and the like. Moreover, in addition to forming a metal layer over the entire surface on at least one side of the aforementioned electric insulating film, there are also cases in which the layer is formed on part of at least one side of the electric insulating film. The thickness of this metal layer is usually 10 angstroms to 100 microns.

The electric insulating film of the present invention is principally used for flexible printed circuits, cover films of the same, flat cables, integrated circuit chip carrier tape, and the like.

Next, working examples will be cited together with the measurement methods used in the present invention in order to describe the present invention in further detail.

#### Measurement methods

##### (1) Intrinsic viscosity

After 0.6 g of film is dissolved (110°C, four hours) in 100 mL of phenol/1,1,2,2-tetrachloroethane as the solvent, a specific amount of the solution is introduced into an Ubbelohde dilution viscometer, and viscosity is measured by means of the conventional method at 30°C.



(2) Trichloroethylene equilibrium absorption rate at 20°C

A 3 cm × 3 cm square test piece is cut into five pieces and weighed. This weight is designated as initial weight  $W_0$ . After being immersed in 500 mL of trichloroethylene (commercial chemical, used as is) for 15 minutes at 20°C, the pieces are removed and the trichloroethylene that has adhered to the surface is wiped off with filter paper. The sample is immediately weighed. This weight is designated as  $W$ . The equilibrium absorption rate of each sample is calculated using the following formula, and the average value is found.

$$\text{Average absorption rate (\%)} = (W - W_0) / W_0 \times 100$$

(3) Percentage dimensional change at 245°C

Five test pieces with a width of 20 mm and a length of approximately 150 mm are sampled in the longitudinal direction, and markings are made at intervals of approximately 100 mm in the center of each piece. The test pieces are hung vertically and heated in a thermostatic chamber kept at a temperature of  $245^\circ\text{C} \pm 3^\circ\text{C}$  for 30 minutes. Then the test pieces are removed and set aside for 30 minutes at room temperature. The distance between the markings is measured, the percentage dimensional change is calculated from the following formula, and the mean value is found.

$$\text{Percentage dimensional change (\%)} = |L - L_0| / L_0 \times 100$$

Here,  $L_0$  is the distance between markings before heating (mm),  $L$  is the distance between markings after heating (mm), and  $| \quad |$  is the absolute value.

(4) Breaking strength

Breaking strength was measured in accordance with ASTM D822-67 at a temperature of 20°C and a humidity of 65% RH. The breaking strength was measured at a test length of 20 mm and a tensile speed of 10 mm/minute.

**Working Example 1**

Polyethylene terephthalate resin with an intrinsic viscosity of 0.75 dL/g was melted at 280°C and extruded from a T-die. The product was quenched on a chill roll at 60°C and then taken up, yielding an amorphous transparent film with a thickness of 80  $\mu$ . This film was ran

through a hot air drying oven while being tented approximately 3%, and was then heat treated for 1 minute at 170°C to obtain an essentially unoriented crystallized film. The intrinsic viscosity of the resulting film was 0.70 dL/g, the trichloroethylene absorption rate was 0.1%, and the percentage dimensional change at 245°C was 0.7% in the longitudinal direction and 0.2% in the traverse direction.

#### Adhesion

A methyl ethyl ketone solution of Toyobo Pyron 300/Millionate MR (solids weight ratio: 80/20) was applied as an adhesive to one side of the aforementioned film and dried at 100°C (adhesive thickness of 20  $\mu$ ). Then 35- $\mu$ -thick electrolytic copper foil was pressed and layered on the adhesive surface with the aid of a 150°C roll to make a copper-clad film.

#### Circuit processing

An etching resist was screen-printed on the copper foil of the copper-clad film that had been fabricated as described above, and was then cured and etched in accordance with JIS C 6481. The product was immersed in trichlene to remove the etching resist and to form a circuit.

#### Solder test

The flexible printed circuit fabricated as described above was treated with a post-flux and then floated for 20 seconds in a solder bath at 250°C with the circuit side facing down. The circuit was checked for floating, swelling, peeling, curling, cracking, and the like of the copper foil.

There were no problems with adhesion or soldering tests. The results are summarized in Table 1.

#### **Working Example 2**

Polyethylene terephthalate with an intrinsic viscosity of 1.3 dL/g was extruded at 290°C, and an essentially unoriented crystallized film (thickness of 80  $\mu$ ) was then fabricated as described in Working Example 1. The evaluation results are shown in Table 1.

### **Working Example 3**

A polyethylene/cyclohexane dimethylene terephthalate (90/10) resin (*trans/cis* ratio of the cyclohexanedimethanol in the starting materials for resin production: 70/30) with an intrinsic viscosity of 0.75 dL/g was extruded at 320°C and crystallized at 230°C as described in Working Example 1 in order to obtain an essentially unoriented crystallized film with a thickness of 100  $\mu$ . The results are shown in Table 1.

### **Comparative Example 1**

Adhesion and soldering tests were performed as described in Working Example 1 using commercial polyethylene terephthalate film (Toray Co., Ltd.: S-10 100  $\mu$ ). The results are shown in Table 1.

### **Comparative Example 2**

Other than the fact that crystallization was not performed, polyethylene terephthalate with an intrinsic viscosity of 0.75 dL/g was treated as described in Working Example 1 in order to obtain an essentially unoriented amorphous film with an intrinsic viscosity of 0.70 dL/g and a thickness of 65  $\mu$ . The results are shown in Table 1.

### **Comparative Example 3**

Other than the fact that crystallization was not performed, polyethylene terephthalate with an intrinsic viscosity of 0.45 dL/g was treated as described in Working Example 1 in order to obtain an essentially unoriented amorphous film with an intrinsic viscosity of 0.42 dL/g and thickness of 65  $\mu$ . The results are shown in Table 1.

**Table 1**

	Film intrinsic viscosity (dL/g)	Trichloroethylene sorption rate (%)	Percentage dimensional changes (%) Longitudinal/transverse	Film (transverse) breaking strength (kg/mm <sup>2</sup> )	Adhesion performance	Soldering performance
Working Example 1	0.70	0.1	0.7/0.2	8.7	Good	Good
Working Example 2	1.0	0.3	1.3/0.1	11.0	Good	Good
Working Example 3	0.62	2.1	2.5/0.6	7.5	Good	Good
Comparative Example 1	0.59	0.1	8.0/4.5	25.0	Good	Impossible
Comparative Example 2	0.70	27.2	Considerable deformation (5% or greater)	7.9	Impossible	Impossible
Comparative Example 3	0.42	28.0	Considerable deformation (5% or greater)	5.3	Impossible	Impossible